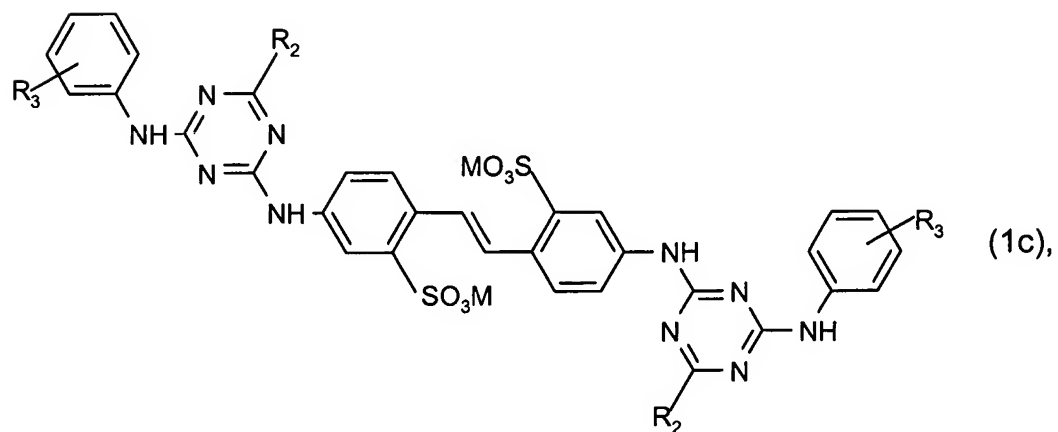
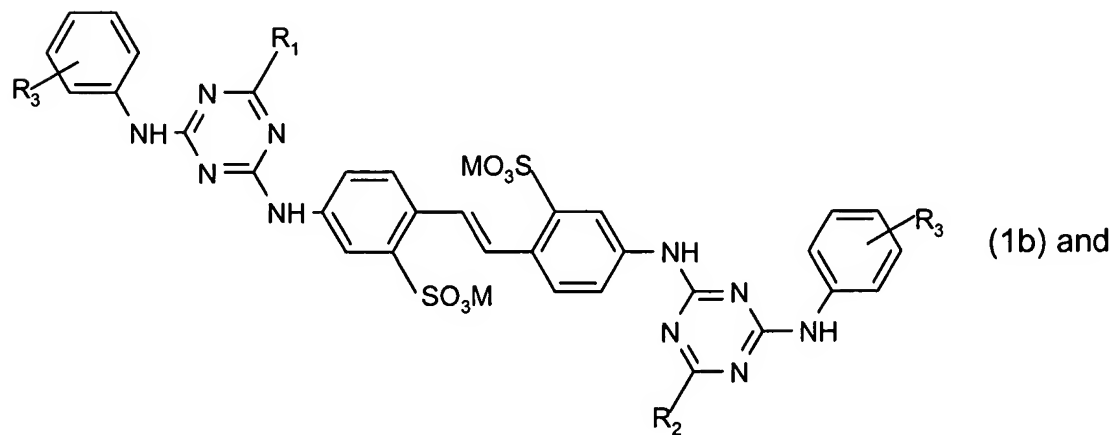
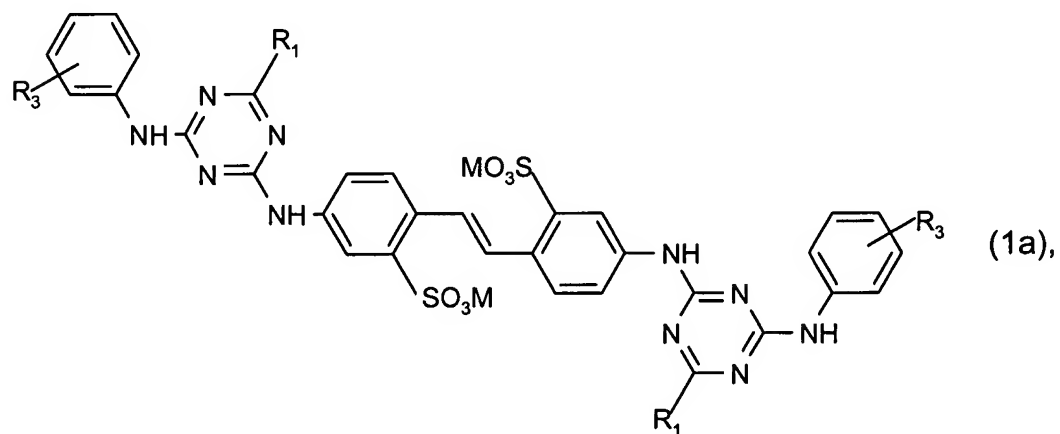


Claims

1-17 (cancelled)

18. (new) A fluorescent whitening agent which is a composition comprising a mixture of two symmetrical compounds (1a) and 1(c) and one asymmetrical compound 1(b) of the formulae



in which R₁ and R₂ are different and

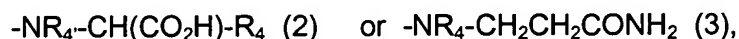
R₁ represents -NH₂, -NHC₁-C₄alkyl, -N(C₁-C₄alkyl)₂, -NHC₂-C₄ hydroxyalkyl, -N(C₂-C₄hydroxyalkyl)₂, -N(C₁-C₄alkyl)(C₂-C₄ hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue,

R₂ represents -NHC₂-C₄ hydroxyalkyl, -N(C₂-C₄hydroxyalkyl)₂, -N(C₁-C₄alkyl)(C₂-C₄ hydroxyalkyl), or an amino acid or an amino acid amide residue from which a hydrogen has been removed from the amino group,

each R₃, independently, represents hydrogen, C₁-C₄alkyl or C₁-C₄alkoxy and

M represents hydrogen, an alkali metal atom, ammonium or a cation formed from an amine.

19. (new) A composition according to claim 18, in which R₂ is an aliphatic amino acid or an amino acid amide residue of the formula



in which each R₄ and R₄, independently, represent hydrogen or a group having the formula -CHR₅R₆ in which R₅ and R₆, independently, are hydrogen or C₁-C₄alkyl optionally substituted by one or two substituents selected from the group consisting of hydroxy, thio, methylthio, amino, carboxy, sulfo, phenyl, 4-hydroxyphenyl, 3,5-diiodo-4-hydroxyphenyl, β-indolyl, β-imidazolyl and NH=C(NH₂)NH-.

20. (new) A composition according to claim 18, in which R₂ is derived from glycine, alanine, sarcosine, serine, cysteine, phenylalanine, tyrosine (4-hydroxyphenylalanine), diiodotyrosine, tryptophan (β-indolylalanine), histidine (β-imidazolylalanine), α-aminobutyric acid, methionine, valine (α-aminoisovaleric acid), norvaline, leucine (α-aminoisocaproic acid), isoleucine (α-amino-β-methylvaleric acid), norleucine (α-amino-n-caproic acid), arginine, ornithine (α,δ-diaminovaleric acid), lysine (α,ε-diaminocaproic acid), aspartic acid (aminosuccinic acid), glutamic acid (α-aminoglutaric acid), threonine, hydroxyglutamic acid, taurine, mixtures and optical isomers thereof, or from iminodiacetic acid or from N-(propionamido)-N-(2-hydroxyethyl)amine.

21. (new) A composition according to claim 18 in which R₂ represent a mono-(2-hydroxyethyl)amino, a di-(2-hydroxyethyl)amino, a di-(2-hydroxypropyl)amino, an N-(2-hydroxyethyl)-N-methylamino, an aspartic acid or an iminodiacetic acid residue.

22. (new) A composition according to claim 21 in which R_2 represents an aspartic acid, or an iminodiacetic acid residue.

23. (new) A composition according to claim 18, in which M represents hydrogen, lithium, potassium, sodium, ammonium, mono-, di-, tri- or tetra- C_1 - C_4 alkylammonium, mono-, di- or tri- C_1 - C_4 hydroxyalkylammonium or ammonium that is di- or tri-substituted with a mixture of C_1 - C_4 alkyl and C_1 - C_4 hydroxyalkyl groups.

24. (new) A composition according to claim 23, in which M represents hydrogen, potassium or sodium.

25. (new) A process for the preparation of the compound mixture of formulae (1a), (1b) and (1c) of claim 18 which process comprises reacting, under known reaction conditions, cyanuric chloride, successively, in any desired sequence, with each of

- i) 4,4'-diaminostilbene-2,2'-disulphonic acid,
 - ii) aniline or aniline substituted by C_1 - C_4 alkyl or C_1 - C_4 alkoxy,
 - iii) an amino compound R_1H and
 - iv) an amino compound R_2H
- or, alternatively
- i) 4,4'-diaminostilbene-2,2'-disulphonic acid,
 - ii) aniline or aniline substituted by C_1 - C_4 alkyl or C_1 - C_4 alkoxy, and
 - iii) a mixture of an amino compound R_1H and an amino compound R_2H

wherein R_1 and R_2 are different and

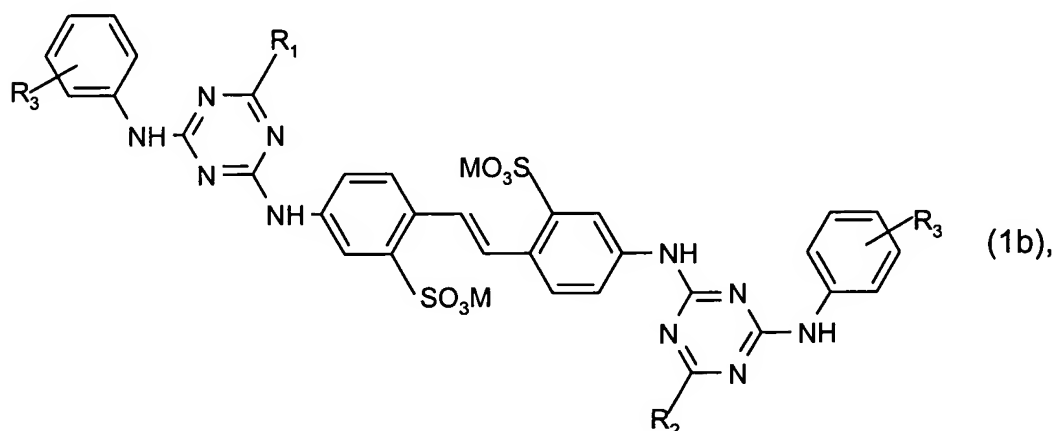
R_1 represents $-NH_2$, $-NHC_1-C_4alkyl$, $-N(C_1-C_4alkyl)_2$, $-NHC_2-C_4hydroxyalkyl$, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4hydroxyalkyl)$, a morpholino, piperidino or pyrrolidino residue and

R_2 represents $-NHC_2-C_4hydroxyalkyl$, $-N(C_2-C_4hydroxyalkyl)_2$, $-N(C_1-C_4alkyl)(C_2-C_4hydroxyalkyl)$, or an amino acid or an amino acid amide residue from which a hydrogen has been removed from the amino group.

26. (new) A process according to claim 25, wherein cyanuric chloride is initially reacted with 4,4'-diaminostilbene-2,2'-disulphonic acid.

27. (new) A process according to claim 26, wherein cyanuric chloride is initially reacted with 4,4'-diaminostilbene-2,2'-disulphonic acid, followed by reaction with aniline or aniline substituted by C₁-C₄alkyl or C₁-C₄alkoxy and then with a mixture of amino compounds R₁H and R₂H.

28. (new) A compound of the formula



in which

R₁ and R₂ are different and

R₁ represents -NH₂, -NHC₁-C₄alkyl, -N(C₁-C₄alkyl)₂, -NHC₂-C₄hydroxyalkyl, -N(C₂-C₄hydroxyalkyl)₂, -N(C₁-C₄alkyl)(C₂-C₄hydroxyalkyl), a morpholino, piperidino or pyrrolidino residue,

R₂ represents an amino acid or an amino acid amide residue from which a hydrogen has been removed from the amino group,

R₃ represents hydrogen, C₁-C₄alkyl or C₁-C₄alkoxy and

M represents hydrogen, an alkali metal atom, ammonium or a cation formed from an amine.

29. (new) A compound according to claim 28, in which R₂ is derived from glycine, alanine, sarcosine, serine, cysteine, phenylalanine, tyrosine (4-hydroxyphenylalanine), diiodotyrosine, tryptophan (β-indolylalanine), histidine (β-imidazolylalanine), α-aminobutyric acid, methionine, valine (α-aminoisovaleric acid), norvaline, leucine (α-aminoisocaproic acid), isoleucine (α-amino-β-methylvaleric

acid), norleucine (α -amino-n-caproic acid), arginine, ornithine (α,δ -diaminovaleric acid), lysine (α,ϵ -diaminocaproic acid), aspartic acid (aminosuccinic acid), glutamic acid (α -aminoglutaric acid), threonine, hydroxyglutamic acid and taurine, as well as mixtures and optical isomers thereof, or from iminodiacetic acid or from N-(propionamido)-N-(2-hydroxyethyl)amine.

30. (new) A composition according to claim 28 in which R_2 represents an aspartic acid, or an iminodiacetic acid residue.

31. (new) A composition according to claim 28, in which M represents hydrogen, lithium, potassium, sodium, ammonium, mono-, di-, tri- or tetra- C_1 - C_4 alkylammonium, mono-, di- or tri- C_1 - C_4 hydroxyalkylammonium or ammonium that is di- or tri-substituted with a mixture of C_1 - C_4 alkyl and C_1 - C_4 hydroxyalkyl groups.

32. (new) A composition according to claim 31, in which M represents hydrogen, potassium or sodium.

33. (new) A composition for whitening synthetic or natural organic materials, which composition contains water, a fluorescent whitening agent comprising a mixture of the compounds (1a), (1b) and (1c), according to claim 18, and, optionally, one or more auxiliaries selected from the group consisting of dispersants, water retention aids, biocides and adjuvants.

34. (new) A method for adding optical brightening agents to paper which method comprises the step of applying a composition of claim 33 either to a paper substrate in a pulp mass, to a paper substrate in a size-press, to a paper substrate in a metering press or contacting a paper surface with a coating application comprising a composition of claim 11.

35. (new) A method, for increasing the Sun Protection Factor (SPF) rating or for the fluorescent whitening of a textile fibre material which method comprises the step of treating said textile fibre material with a composition of claim 33.